Ambipolar gate effect and low temperature magnetoresistance of ultrathin $La_{0.8}Ca_{0.2}MnO_3$ Films

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Ultrathin $La_{0.8}Ca_{0.2}MnO_3$ films have been measured in a field-effect geometry. The electric field due to the gate produces a large ambipolar decrease in resistance at low temperatures. This is attributed to the development of a pseudogap in the density of states and the coupling of localized charge to strain. The gate effect and magnetoresitance are interpreted in a consistent framework. The implications for the low temperature behavior of a manganite film in the two dimensional limit are discussed.

Ultrathin films of manganites present a unique opportunity for studying the low temperature properties of a half-metallic fully spin polarized electron gas in the two-dimensional limit, with tunable disorder. In the past decade, there has been a resurgence of interest in the possible ground states of highly correlated two-dimensional electron gases (2DEGs) [1], especially regarding the existence of a metallic state at low temperatures. Both high mobility 2DEGs and highly disordered amorphous ultrathin metal films exhibit large positive magneto-resistance for in-plane magnetic fields at low temperatures [2]. This is consistent with theoretical arguments that attribute the existence of a metallic ground state to strong antiferromagnetic or singlet correlations between electrons [3] in the appropriate range of densities. When these correlations are quenched by the Zeeman energy in a magnetic field, the metallic state is lost, and the resulting insulator accounts for the large positive magnetoresistance. In the manganites, due to strong Hund's coupling between the core Mn spins and the conduction electrons, a fully spin-polarized electron gas is obtained at low temperatures in the ferromagnetic state. This does not allow for singlet correlations, and should result in an insulating ground state in the two-dimensional limit, even in the presence of very weak disorder. Furthermore, the strong cross-couplings between strain, charge and spin that exist in these systems make for unique features in their response to external perturbations. Localized electrons at the Mn^{3+} sites cause lattice distortions via the Jahn-Teller effect, and as a result, strain fields develop within the lattice. When electrons are delocalized, either by better alignment of spins by an external magnetic field (double-exchange mechanism) or via changing the local density of charge (electrostatic gating), the strain is relieved and the resulting disorder potential is reduced.

In this letter we investigate the low temperature properties of ultrathin films of $La_{0.8}Ca_{0.2}MnO_3$ (LCMO) to gate and magnetic fields. This composition of LCMO is close to the phase boundary between a ferromagnetic metal (FM) at higher Ca doping and a ferromagnetic charge ordered insulator (F-COI) at lower doping. Bulk single crystals of similar composition are believed to exist in a mixed phase with coexisting regions of insulating and metallic properties [4], with the behavior at low temperatures arising from percolation of metallic regions in the material. The samples are typically 21 u.c ($\backsim$82Å) thick films of $La_{0.8}Ca_{0.2}MnO_3$ grown using ozone-assisted molecular beam epitaxy on surface treated $SrTiO_3$ substrates that have been thinned to 35-50μm locally [5] permitting configuration in a field-effect geometry. A Pt electrode (1000Å thick) deposited on the back of this thinned region serves as the gate. The as-grown manganite film is patterned into a wire of 100μm width, with tabs for carrying out four terminal measurements. All transport measurements reported here were performed using standard DC techniques. The gate-drain current was always monitored, and remained below 0.6nA for gate electric fields as high as 85kV/cm, while typically the source-drain measurement current was 100nA.

We observe a magnetic transition at about 150K with an accompanying resistive transition at about 138K (T_c), from an activated insulating state to a nominally metallic state, along with CMR [Fig.1(a)]. However, at the lowest temperatures (<36K) there is a re-entrant insulating phase [6]. Near the resulting minimum, the film resistance has a large ambipolar susceptibility to a gate electric field along with a large magnetoresistance, in contrast to earlier work on thicker films [7] where the largest susceptibilities were observed only near the insulator-metal transition at high temperature. We propose that the ambipolar gate response is due to the opening up of a pseudogap in the density of states (DOS) at low temperatures. The large magnitude of the response to gate and magnetic fields is due to the motion of domain boundaries separating insulating and metallic phases. Glassy dynamics have also been observed in the response of these films to external perturbations [8], attributed to the relaxation of strain via cross-couplings to spin and charge.

The transition from the low temperature metal to the re-entrant insulator at the lowest temperatures is continuous. This is borne out by the absence of any hysteresis upon cooling and then warming the sample, for all

temperatures below 30K. There is hysteresis above this temperature, associated with the insulator-metal transition at T_c, as seen in other work [9]. We interpret the hysteretic behavior as a signature of the nucleation (melting) of metallic patches upon cooling (warming) near T_c. Upon cooling to low temperatures, these patches form an infinite cluster and metallic behavior ensues. Within that framework, the lack of hysteresis in the transition into the re-entrant insulator indicates that the insulator/metallic fraction does not change upon entering this phase. However, the degree of disorder (strain) within the insulating patches, and the resultant martensitic accommodation strain may become higher, as has been observed in optical images at low temperatures [10].

It is known from photoemission [11] and optical conductivity [12] experiments that the density of states (DOS) at the Fermi level in the manganites is severely depleted, presumably due to the opening of a pseudogap [13] or a minimum in the DOS at the chemical potential. This has also been observed in scanning tunneling spectroscopy at low temperatures in the mixed phase [14]. A pseudogap is predicted in the mixed phase in two dimensions in the presence of either structural [15] or spin disorder [16]. The minimum is most pronounced at low temperatures, and is eventually washed out as the temperature is raised. A pseudogap has been observed in photoemission experiments on layered manganites [13], with an accompanying upturn in the resistance at the lowest temperatures.

We studied the response of the film to an applied gate field in the temperature range from 2K to 138K [Fig.2(a)]. The most striking feature of the data is that the response is ambipolar for nearly the whole temperature range. At T_c, the resistance decreases for negative gate voltages (which induce hole type carriers), but there is no change in resistance for positive gate voltages. Upon lowering the temperature, the response to a positive gate voltage grows, although there is a larger effect for negative gate voltages than for positive. The ambipolar nature of the response may suggest electrostriction in the STO being the cause, but several results refute this. LCMO thin films exhibit an increase in resistance, upon an increase in the strain (positive sign for biaxial expansion) [17]. The maximum strain due to electrostriction [5] is about $6\text{x}10^{-4}$, almost an order of magnitude less than the strain due to the lattice mismatch between STO and LCMO, which is $7.7\text{x}10^{-3}$. The positive sign of the strain indicates that electrostriction would increase the mismatch between LCMO and STO, increasing the resistance further, and not decreasing it as we observe. Additionally, STO exhibits no appreciable electrostriction effects below electric fields of 2kV/cm, and the effect saturates at electric fields above 15kV/cm [5] at low temperatures. We find electric fields as low as 0.29 kV/cm reduce the resistance, and there is no evidence of saturation of the gate effect for electric fields as high as 85kV/cm. Furthermore, using an identical dielectric, non-ambipolar modulation of the transition temperature has been obtained for ultrathin $NdBa_2Cu_3O_{7-\delta}$ films in a field effect geometry [18], with enhancement (reduction) of T_c with a negative (positive) gate voltage. Thus is likely that the ambipolar effect reflects the electronic properties of the ultrathin LCMO.

An ambipolar gate effect can be signature of a minimum in the density of states. In amorphous covalent semiconductors, this occurs due to tails in the DOS in the mobility gap [19]. In a two-dimensional disordered system, the evolution from a metal to an insulator (e.g. as a function of decreasing thickness) may be accompanied by the opening of a correlation gap in the DOS at the Fermi level [20]. Normally, one would not expect an ambipolar gate effect if the minimum in the DOS tracks the chemical potential. However, if the relaxation time for charge to equilibrate is longer than the measurement time, turning on a gate voltage abruptly can lead to a non-ergodic distribution of charge, where the chemical potential rides up the DOS about the minimum for both signs of applied gate voltage, giving rise to an ambipolar enhancement of conductance. This is usually accompanied by glassy behavior in the charge relaxation [21].

Our observations may be explained by the physics of the mixed phase at low temperatures. The strongly insulating regions would be largely transparent to the gate electric field, and remain unaffected. The strongly conducting regions would have a DOS consistent with the dopant concentration, and would also be weakly affected, since the electric field would be screened out in the first few unit cells. The boundaries of these regions would have intermediate DOS and would couple maximally. Because of strong coupling between localized charge and local strain via the Jahn-Teller effect, any change in the charge configuration in these boundary regions causes a corresponding change in the strain. Competing strain fields in the material gives rise to a metastable energy landscape with hierarchical energy barriers for relieving the strain [22]. This causes a hierarchical and glass like response of the system to any external force, due to either gate or magnetic field, that seeks to change the phase boundaries between the insulating and conducting regions. The hierarchy of energy barriers being crossed in either case is the same since they are governed by the same strain field [8]. If the DOS has a minimum, and the electronic response is glass like, the conductance of these boundary regions will be enhanced for both signs of gate voltage. The resulting delocalized charge causes the strain fields to be relieved and the phase fraction of the metallic region increases at the expense of the insulating region. Domain boundaries move irreversibly through a metastable pinning landscape, and the system latches the change caused in the conductance by the gate.

We first comment on the qualitative nature of the gate effect and its temperature dependence. We use the ratio of the magnitude of the gate effect for positive and negative voltages as a measure of ambipolarity, independent of the magnitude of the effect. Upon lowering the temperature below T_c, this measure is seen to increase

and then level off [Fig.2(b)] at the lowest temperatures, nominally consistent with the predicted evolution of the pseudogap [15,16]. The pseudogap is likely suppressed by the application of a magnetic field, since this reduces the disorder and makes the sample becomes more homogeneously metallic. This is borne out by the suppression of the upturn by application of a magnetic field [Fig.2(c)].

Next, we turn to the magnitude of the effect. In understanding the response, we have to consider the nonlinear and temperature dependent dielectric constant of STO [5], and the susceptibility of the film. The maximum response to electric field occurs in the region near the onset of the reentrant insulating state. Although the response is greatest at 30K, the response at 50K is comparable when considering that dielectric constant of STO [5] at 50K is less than at 30K and thus the equivalent charge transfer at that temperature will be less. However, at 2K, when the induced charge is maximal ($\backsim$5.8x10^{13}cm^{-2} for 40kV/cm) the response is not as large as at 50 K where the charge transfer is significantly less ($\backsim$4.4x10^{13}cm^{-2} for 40kV/cm). Other manganite films with re-entrant insulating behavior have also shown a reduction in the gate effect at temperatures below 50K [23]. Also, looking at the magnetoresistance data, the resistance of the manganite film is more susceptible to the initial application of a magnetic field at 30K than at 2K [Fig.3(a)] We consider this in the framework of pinned domain walls separating insulating and metallic regimes, where the hierarchy of pinning is determined by strain fields, which can be coupled to both by magnetic and electric fields [8], one would expect the magnitude of the response to gate electric field and magnetic field to exhibit a similar temperature dependent response. At 30K, the gate effect is suppressed magnetic field [Fig.3(b)]. In zero magnetic field, the gate effect at 2K is less than at 30K [Fig.2(a)]. However, when 9T is applied to the film and then a gate is turned on, the gate effect is larger at 2K than at 30K [Fig 3(c)]. All of this can be understood in the framework of a 'general susceptibility', describing the ease with which a domain boundary moves in a hierarchical pinning landscape. Since magnetic and gate electric fields are 'equivalent' in the response of the film in the mixed phase, we use the magnetoresistance as a measure of this susceptibility. The magnetoresistance as a function of field [Fig.3(a)] clearly shows that the slope is steeper at 30K than at 2K for low fields. However at higher fields, this situation is reversed. Thus, turning on a small gate field in the presence of a magnetic field would have the effect of taking a differential measurement of the MR, and this explains the difference between the 2K and 30K data in the presence and absence of a magnetic field.

A number of reports have been made of a field-direction independent response to applied gate electric fields in manganite thin films [7,24]. One model for the ambipolar response proposes that the applied electric field affects the lattice distortions in the manganite, causing corresponding changes in resistivity [24]. In this picture, with the applied electric field acting as a perturbation to the Jahn-Teller distortion within the manganite, it is unclear why the ambipolar response would have the temperature dependence that we see. Indeed, the original proposal was used to explain ambipolar behavior in the vicinity of and above the temperature of the peak resistance.

Another alternative mechanism that does not involve the density of states directly is due to just the electrostatic forces felt by the accumulated charges in the metallic regions. As mentioned earlier, the insulating regions are transparent to the gate electric field, and some of these field lines would terminate on metallic regions at the boundaries with the insulating regions. These charges would then experience a force that would literally pull the boundaries further into the insulating regions, increasing the fraction of metallic phase. The same arguments regarding the hierarchy of barriers surpassed would hold. However, in a purely hole doped system, there would also be accumulation and depletion at the boundaries due to negative and positive gate voltages, and it has been argued that this gives rise to a non-ambipolar effect [7]. The temperature dependence of the symmetry of such an effect is also not obvious.

In conclusion, the nature and magnitude of the gate effect, and the relation to magnetoresistance are consistently explained within a mixed phase scenario, invoking a pseudogap and a hierarchical metastable energy landscape for the motion of the phase boundaries. The absence of an ambipolar effect at higher temperatures is likely due to a greater homogeneity of phases in this regime. The low dimensionality of our films may also be crucial in observing the effects. The possibility of a field (disorder) induced insulator-metal transition in the re-entrant insulator with the existence of a quantum critical point will be the subject of future work.

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[1] E. Abrahams, S.V. Kravchenko and M.P. Sarachik, Rev. Mod. Phys. **73**, 251 (2001).

[2] K.M. Mertes et al., Phys. Rev. B **60**, R5093 (1999); L.M. Hernandez, A. Bhattacharya, K.A. Parendo and A.M. Goldman, Phys. Rev. Lett. **91**, 126801 (2003).

[3] P. Phillips and D. Dalidovich, Science **302**, 243 (2003); S. Chakravarty, S. Kivelson, C. Nayak and K. Voelker, Philos. Mag. B **79**, 859 (1999).

[4] P.A. Algarabel *et al.*, Phys. Rev. B **67**, 134402 (2003).

[5] A. Bhattacharya, M. Eblen-Zayas, N. Staley, W.H. Huber and A.M. Goldman, accepted for publication in Appl. Phys. Lett. (2004).

[6] M. Ziese *et al.*, J. Appl. Phys. **91**, 9930 (2002); M. Bibes

et al., Phys. Rev. B **66**, 134416 (2002).
[7] H. Tabata and T. Kawai, IEICE Trans. Electron. **E80-C** , 918 (1997); T. Wu *et. al.*, Phys. Rev. Lett. **86**, 5998 (2001).
[8] A. Bhattacharya, M. Eblen-Zayas, N. Staley, A. L. Kobrinski and A.M. Goldman, cond-mat/0407607.
[9] X. J. Chen, H.-U. Habermeier, and C. C. Almasan, Phys. Rev. B **68**, 132407 (2003).
[10] V. Podzorov *et al.*, Phys. Rev. B **64**, R140406 (2001).
[11] J.H. Park *et al.*, Nature **392**, 794 (1998); D.D. Sarma *et al.*, Phys. Rev B **53**,6873 (1996).
[12] Y. Okimoto *et al.*, Phys. Rev. B **55**, 4206 (1997).
[13] T. Saitoh *et al.*, Phys. Rev. B **62**,1039 (2000); Y.-D. Chuang *et al.*, Science **292**, 1509 (2001).
[14] M. Fath *et al.*, Science **285**, 1540 (1999); Ch. Renner *et al.*, Nature **416**, 518 (2002).
[15] A Moreo, S. Yunoki and E. Dagotto, Phys. Rev. Lett. **83**, 2773 (1999).
[16] S. Kumar and P. Majumdar, Phys. Rev. Lett. **92**, 126602 (2004).
[17] R.A. Rao *et. al.*, Appl. Phys. Lett. **73**, 3294 (1998); J.Z. Sun *et. al.*, Appl. Phys. Lett. **74**, 3017 (1999).
[18] D. Matthey, S. Gariglio and J.-M. Triscone, Appl. Phys. Lett. **83**, 3758 (2003).
[19] D.F. Barbe, J. Vac. Sci. Tech. **8**, 102 (1971).
[20] V.Yu. Butko, J.F. DiTusa and P.W. Adams, Phys. Rev. Lett. 84, 1543 (2000).
[21] Clare C. Yu, Phys. Rev. Lett. **82**, 4074 (1999); J.H. Davies, P.A. Lee and T.M. Rice, Phys. Rev. Lett. **49**, 758 (1982).
[22] K.H. Ahn, T. Lookman and A.R. Bishop, Nature **428**, 401 (2004).
[23] I. Pallecchi et. al., J. Appl. Phys. 95, 8079 (2004).
[24] S.B. Ogale *et. al.*, Phys. Rev. Lett. **77**, 1159 (1996).

FIG. 1. Resistive and magnetic transitions for 21 u.c. thick LCMO film and (inset), CMR and resistive upturn for the sample at low temperatures.

FIG. 2. Ambipolar gate effect and the pseudogap. (a) Temperature dependence of the gate effect. Note, this does not account for the temperature and field dependence of the dielectric properties. (b) Temperature dependence of the 'ambipolarity' or symmetry of the gate response. (c) Dependence of the re-entrant phase on applied magnetic field (different sample).

FIG. 3. Susceptibility to external fields. (a) Magnetoresistance for in-plane fields. The data at 2K and 30K have greater density of points. (b) Gate effect at different magnetic fields at 30K. (c) Gate effect at 2K and 30K, with 9T magnetic field on.

FIG. 1. M. Eblen-Zayas et al.

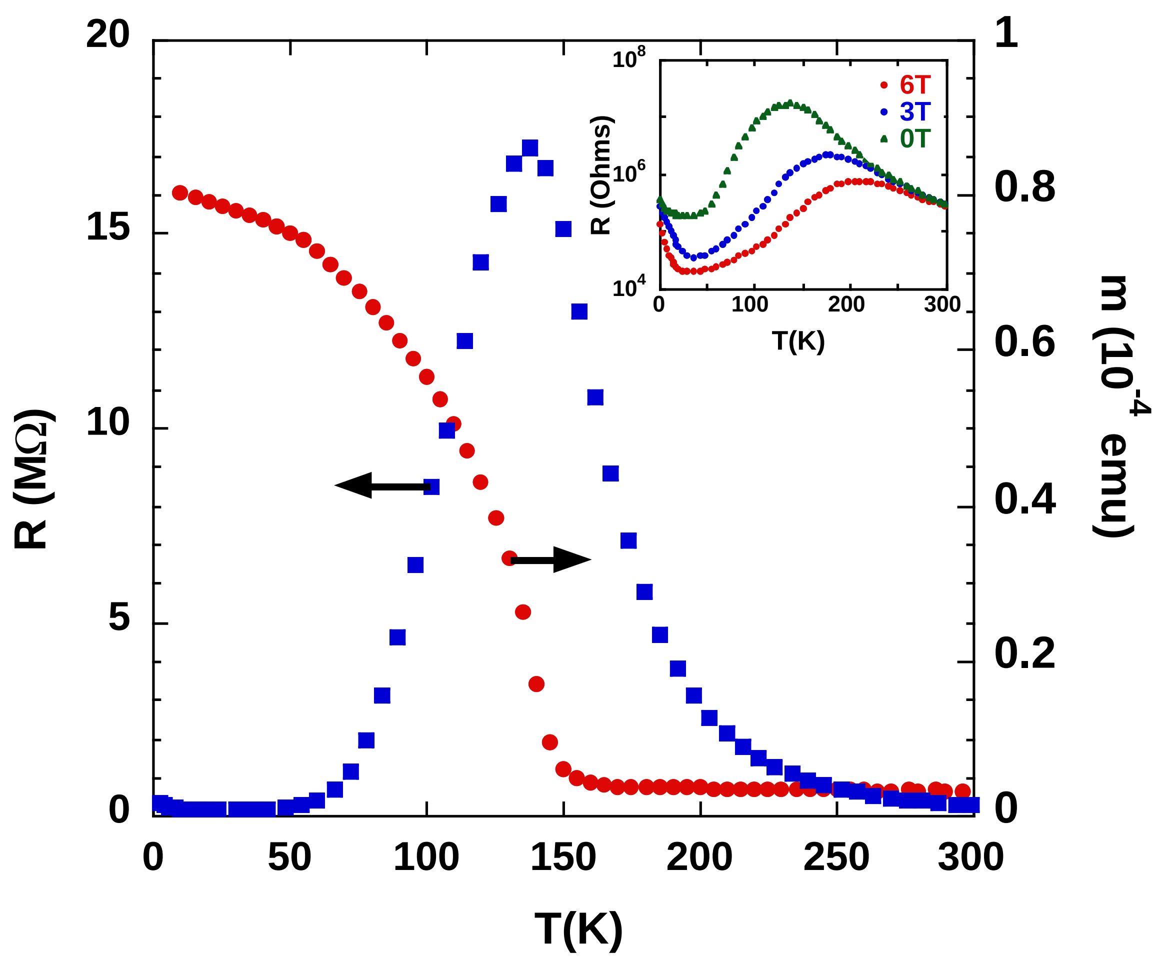

FIG. 2. M. Eblen-Zayas et. al.

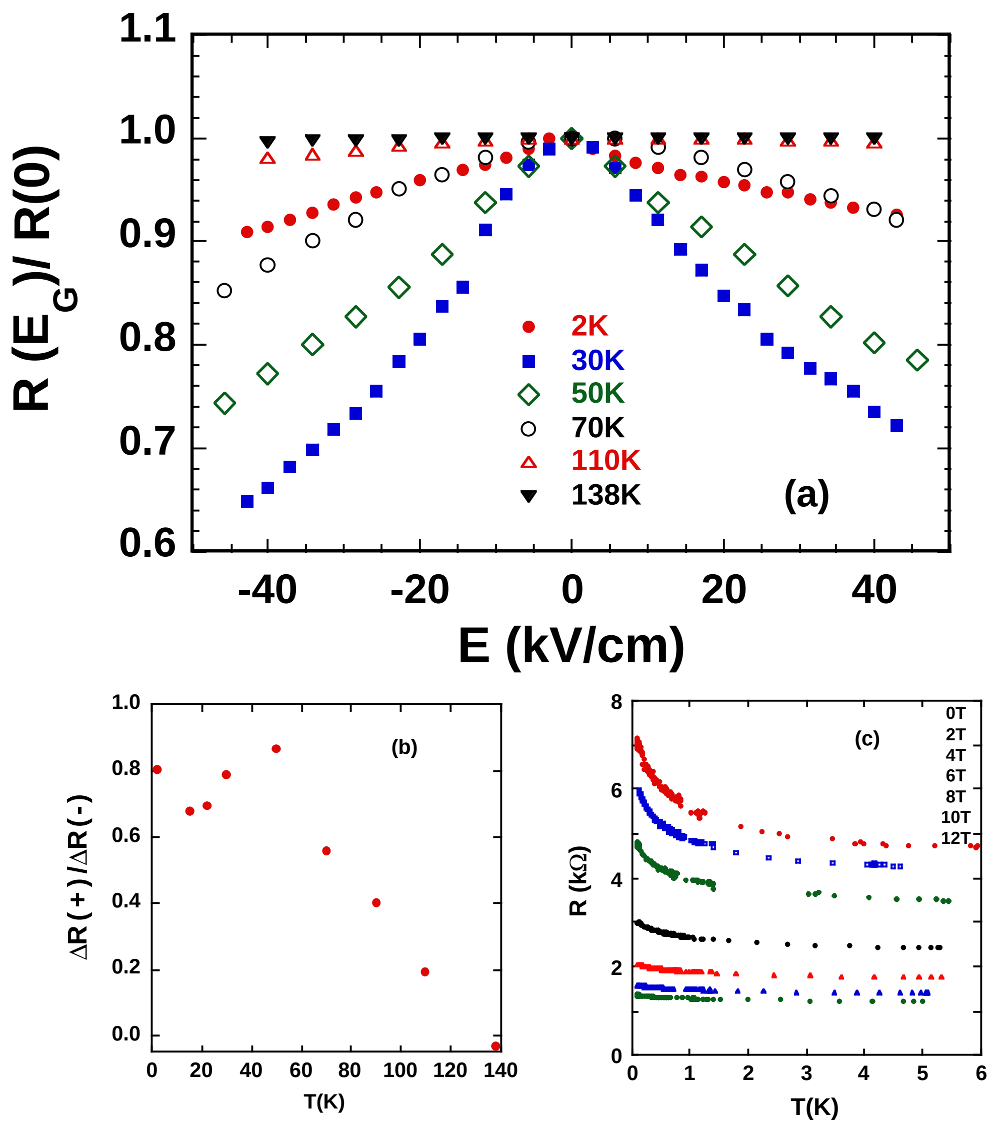

FIG. 3. M. Eblen-Zayas et. al.